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PARTICLE GROWTH PROCESSES IN SCREENING SMOKES.(U)
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PARTICLE GROWTH PROCESSES IN SCREENING SMOKES.

FINAL REPORT.

J. R. BROCK

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The view, opinions, and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation. This study was sponsored by the Army Smoke Research Program, Chemical Systems Laboratory, Aberdeen Proving Ground, MD.

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Experimental and theoretical studies have been carried out on the growth of oil condensation aerosols of the type employed by the U.S. Army as screening smokes. Studies of the vapor pressures of the complex oils have been carried out and new methods and results have been obtained. An experimental system has been developed, a laminar coaxial jet, from the vapor of oil condensation aerosols. The growth processes, for complex oil mixtures, are found to lead to the appearance of bimodal number density functions whose development

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20. Abstract (continued)

we have studied for the first time. A numerical model has been developed of the experimental laminar coaxial jet and used in a theoretical study of coagulation in the jet.

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Objectives of Research

Work under this grant was intended to add to the technological base of the U.S. Army's smoke program as well as to contribute to fundamental knowledge of aerosol physics. In this study the principal focus has been on the influence of smoke agent properties and particle growth processes on the particle size and composition distributions, which of course dictate the obscuration efficiency of a smoke plume/puff.

Summary of Results

This research has dealt principally with liquid oil aerosols of the type employed by the U.S. Army as oil smokes. The approach has been primarily experimental with, however, important contributions from theoretical analysis in gaining a better understanding of aerosol growth processes in plumes.

This summary gives the important findings of our research into the processes involved in aerosol growth in screening smokes. It begins with results obtained in experimental studies of oil evaporation. Next, a description is given of an experimental system developed during this grant. Finally, experimental and theoretical studies with the experimental system are summarized.

Studies of Vapor Pressures of Complex Oil Mixtures

A number of complex oils are in common use by the U.S. Army as obscurant materials. These include the fog oils, SGF-1 and SGF-2, diesel oils, and others. These materials are extremely complex and consist of many thousands or hundreds of thousands of different chemical species. Any investigation of the formation and growth of these oil fogs must first consider the meaning of the vapor pressure of such complex mixtures and the role of this complexity in the resultant particle size distribution.

(1) The piezoelectric microbalance technique.

A new method has been developed and used to study the vapor pressures of complex oil mixtures through measurement of evaporation rates (1). This method uses the frequency shift of a piezoelectric quartz crystal as a deposited oil film evaporates in a vacuum. Reasonable agreement is found between vapor pressure measurements of dioctylphthalate at 25°C by this technique and values reported in the literature. This method has also been used to estimate the vapor pressure of complex oils such as diesel and fog oils.

(2) New theory for evaporation of complex oil mixtures.

An approximate theory has been developed for determination of the dependence of vapor pressure on molecular weight for complex oil mixtures at a given temperature. The theory proceeds from the molecular mass density function, $n(m,t)$, for the very large number of chemical species found in complex oil mixtures. The first two moments of $n(m,t)$ give the total number, $N(t)$, of chemical species in the mixture at time, t:

$$N(t) = \int_0^\infty n(m,t) \cdot dm$$

and the total mass, $M(t)$, of the oil sample:

$$M(t) = \int_0^\infty mn(m,t)\, dm$$

The theory successfully correlates evaporation data obtained from two quite different experimental systems: oil film evaporation *in vacuo* and oil droplet evaporation at near atmospheric pressure. In the full paper, suggestions are given for improvements in the analysis (2).

Experimental Investigations of Formation and Growth of Condensation Aerosols

In most technical applications, condensation aerosols are generated in turbulent host gases. As a first step toward investigation of aerosol evolution in such turbulent systems, we are studying generation of condensation aerosols in the much simpler experimental system represented by a laminar, coaxial jet.

This permits elucidation of the basic aerosol growth mechanisms under mathematically definable conditions. The still unresolved closure problem of turbulence prevents a "first principles" analysis of turbulent systems.

Fig. 1 shows a portion of the experimental system for studying condensation aerosol formation and growth in a laminar, coaxial jet. It shows the arrangement of the clean air supply system, the vapor generator, the laminar, coaxial jet system, and the optical and data acquisition system. Not shown is the Royco optical particle counter and signal processing and computer system used in measurement of particle size distributions in the laminary, coaxial jet by sampling from the jet system.

Initially, condensation aerosol has been studied from the vapors of two substances: glycerol, a pure substance, and fog oil SGF-2, a complex hydrocarbon oil mixture consisting of many thousands of chemical species with molecular weights covering the range 240 - 500, whose properties have been discussed in another section and reported in detail (2). A complete description of our experiments with glycerol and fog oil can be found in a paper now being submitted for publication. These experiments include in situ measurements of mean particle sizes using an argon-ion laser and the Rayleigh-Gans-Debye theory as well as extensive measurements using a sampling probe and Royco optical particle counter and automatic data system. The latter experiments cover a wide range of experimental conditions and agree well with the in situ measurements for glycerol, but not for fog oil owing to possible multiple scattering and appearance of bimodal density functions for this oil.

The notable feature of the fog oil studies are the bimodal number density functions which develop at larger downstream axial distances in the jet. The explanation for such bimodal density functions has been detailed in theoretical studies of condensation growth of aerosols described in another report. Simply

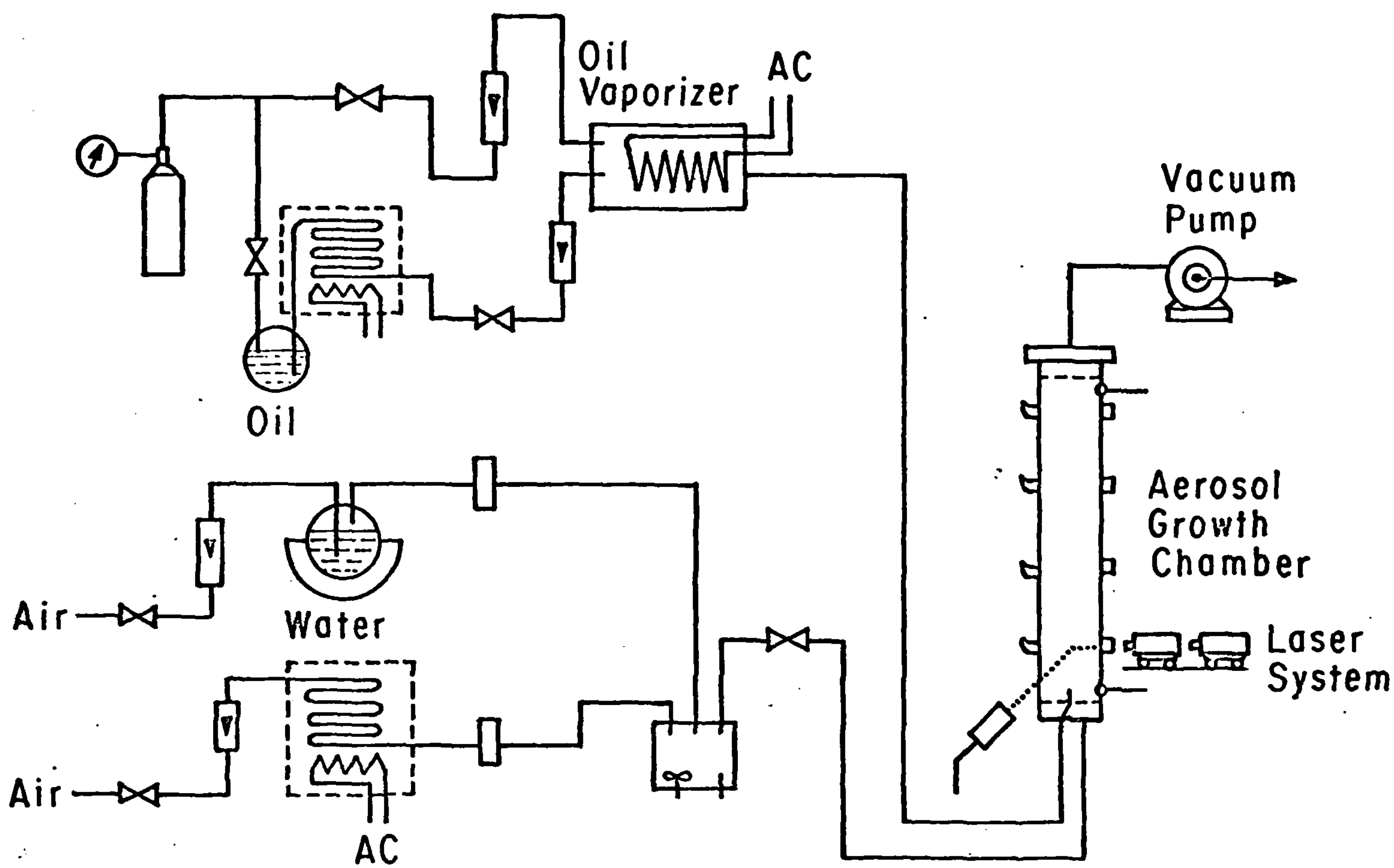

Figure 1. Schematic Diagram of Experimental System for Study of Condensation Aerosols.

stated, the bimodal forms are the result of the initial rapid nucleation of the higher molecular weight fractions of fog oil upon which subsequently the lighter fractions condense. Also of interest is the much larger mean particle diameters achieved with fog oil compared to those for glycerol. The mass mean particle diameter achieved by glycerol at an axial downstream distance of 257 nozzle diameters (nozzle diameter = 1.3 mm) is around 0.5 μm. For fog oil at the same downstream distance, the mass mean diameter is around 1-2 μm. As observed empirically many years ago by Langmuir and Schaeffer and as proved by our theoretical analyses, mean particle size can be greatly increased by causing condensation aerosols to grow by condensation rather than by coagulation. Our experimental results confirm these other findings. A paper giving first results obtained in the laminar coaxial jet system is being submitted for publication (3).

Coagulation in Laminar Coaxial Jets

The evolution of an ultrafine coagulating, coalescing aerosol in a laminar, isothermal coaxial jet has been studied in computer "experiments" as a first step in the investigation of nucleation and growth of aerosols in turbulent jets. These studies are simulations of an experimental system described above and shown schematically in Fig. 1.

For the laminar, isothermal coaxial jet, coagulation is the dominant process affecting the total number concentrations of an initial aerosol with no condensable vapor. At the particle concentration levels studied in these simulations, most of the change in aerosol characteristics occurs within axial distances of the order of 10^2 nozzle diameters. Over the cross section of the aerosol jet, mean particle diameter increases monotonically with downstream distance. However, the standard deviation of the particle number density function increases rapidly close to the nozzle, achieves a maximum, and then decreases. At the periphery of the jet at small downstream distances, the polydispersity of the aerosol and the size dependence of Brownian diffusion of particles lead to a bimodal particle number denstiy function.

Results are also reported of a computer "experiment" for coagulation of an aerosol in a non-isothermal laminar, coaxial jet. For the conditions studied, five processes affect the aerosol particle size distributions in the heated jet. These are axial and radial convection, Brownian coagulation, Brownian diffusion, and thermophoresis. Evolution of the aerosol in the heated jet differs markedly from that found for the same initial aerosol in an isothermal laminar, coaxial jet. Along the axis of the heated jet, marked bimodal number and mass density functions are found at the periphery of the jet near the nozzle. These bimodal functions arise from Brownian diffusion and thermophoresis. As in the isothermal process affecting the particle size distribution, coagulation is the dominant rate process affecting the particle size distribution.

These results are reported in a paper now in press (4).

Publications and Technical Reports

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4. Kajiuchi, T. and Brock, J. R., "Coagulation of an Aerosol in a Laminar Coaxial Jet", In Press (J. Colloid and Interface Sci.).

Participating Scientific Personnel

1. K. S. Yom, Ph.D. Candidate (degree expected, May, 1981).
2. Dr. S. H. Baek (postdoctoral research associate).
3. Dr. T. Kajiuchi (postdoctoral research associate)
4. J. E. Harris, Ph.D. Candidate (degree expected May, 1981).

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